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(18) Applicant: UNIROYAL CHEMICAL COMPANY,  
Inc.  
World Headquarters  
Middlebury Connecticut 06749(US)

(19) Inventor: Wefer, John Michael  
Saddle Ridge Road Newtown  
Fairfield Connecticut 06470(US)

(20) Representative: Harrison, Michael Robert et al  
Urquhart-Dykes & Lord 91 Wimpole Street  
London W1M 8AH(GB)

(54) Impact resistant polyethylene terephthalate/polycarbonate/PE blends.

(57) High impact strength blends based on polyethylene terephthalate resin, polyethylene resin, aromatic polycarbonate resin and styrene-acrylonitrile grafted copolymers of EPDM. This invention is of special value in improving the engineering properties of PET:PE bottle recycle products.

EP 0 322 095 A2

# IMPACT RESISTANT POLYETHYLENE TEREPHTHALATE/ POLYCARBONATE/POLYETHYLENE GRAFT COPOLYMER BLENDS

## FIELD OF THE INVENTION

5 This invention relates to improved high impact compositions comprising blends of polyethylene terephthalate resin, polycarbonate resin, polyethylene and a grafted EPDM copolymer composition. In another aspect, the invention pertains to high impact, polyethylene terephthalate blends derived from the recycling of polyethylene terephthalate bottles together with the polyethylene base cups.

## BACKGROUND OF THE INVENTION

10 Products designed around the thermoplastic polyester resins are in great demand because of the tremendous versatility of these resins and their inherent properties. Polyethylene terephthalate (PET) with its slow crystallization rate has largely been used in packaging; however, nucleating agents have been used to increase the crystallization rate, and post-molding heat treatments (annealing) can be used to crystallize molded articles made of PET. The crystallized PET tends to higher tensile strength, stiffness, and heat resistance compared to uncrystallized PET.

20 Because of the unique engineering properties of the PET resin and in a continuing search to improve the overall property spectrum of molded PET, the resin has often been blended with numerous other resins. Additionally, specific additives have also been incorporated to increase the general level of the impact strength of the PET-containing blends as well as the consistency of the impact properties in molded articles.

25 Illustrative of specific additives or impact modifiers that have been proposed to be used with thermoplastic polyester resins are the family of modifiers comprising polyalkyl acrylates, methacrylates, and/or ethacrylates (Brinkmann et al; U.S. Patent 3,591,659) typical of which is Acryloid KM-330, a polyacrylate resin sold by Rohm & Haas Co.

30 Certain rubber resin graft copolymers have also been proposed to affect impact resistance of polyester resins. In U.S. Patent No. 3,919,353 (Castelnuovo et al) EPDM grafted with common ethylenically unsaturated monomers is disclosed to be an effective impact modifier. Polybutadiene is taught to be the preferred elastomer. All of the examples use polybutadiene grafted with styrene-methyl methacrylates and the elastomer is crosslinked.

35 One of the resin blends preferred for its overall balance of molded properties is that comprising polyesters and polycarbonate (PC) resins. It should be noted that the impact strength at room temperature of most polycarbonates is relatively high. For example, the Notched Izod Impact at Room Temperature (NIRT)(ft-lb/in.) of numerous commercial polycarbonates such as LEXAN 103 (trademark of General Electric) is often in the range of from about 16 to 20 ft-lb/in. However, to obtain maximum chemical resistance of polyester/polycarbonate blends, it is desirable to keep the polycarbonate content low.

40 Furthermore, polycarbonate resin is susceptible to mechanical stress cracking which can be exacerbated by exposure to unfavorable environments such as the presence of solvents or solvent vapors. This tendency to stress crack provides an additional reason to utilize as little polycarbonate resin as possible in these polyester blends.

45 When adding impact modifiers to polyethylene terephthalate/polycarbonate blends, susceptibility to the effects of ester interchange on the final blend properties must also be taken into account. Blends produced under high shear conditions may have unpredictable properties due to reaction (ester interchange) between the polyester and polycarbonate components. The ester interchange reaction can be minimized, of course, by the use of inhibitors and/or by lowering the shear level.

50 Various blends based on polyester resin, polycarbonate resin and graft copolymer rubber-resin materials have heretofore been proposed (e.g., U.S. Patent Nos. 3,591,659, 3,919,353; 4,022,748; 4,034,013; 4,034,016; 4,044,073; 4,096,202; 4,260,690 and 4,280,949.

U.S. Patent No. 4,257,937 teaches the impact modification of polyester/polycarbonate blends via a polyacrylate resin such as Acryloid KM-330 and 7709-XP (trademarks of Rohm & Haas Co.)

U.S. Patent No. 4,172,859 (Epstein) discloses blends of polyesters, polycarbonate and an impact

modifier which can comprise various monomers, combinations of which can include EPDM.

U.S. Patent No. 4,493,921 represents another step in the continuing effort to improve the level of performance of polyester-type resin/polycarbonate-type blends via the addition to polybutylene terephthalate (PBT)/PC blends of an impact modifier comprising an ethylene-propylene-non-conjugated diene (EPDM) rubber spine grafted with either a styrene/acrylonitrile monomer mixture, a styrene/methyl methacrylate monomer mixture or a methyl methacrylate monomer mixture.

U.S. Patent No. 4,547,536 describes the preparation of PET/PE molding resin from soda bottle regrind via the incorporation of a desiccant, mineral oil, and a block copolymer impact modifier. Unfortunately, the blends described by this patent have rather low impact strengths, especially Notched Izod Impact values.

One of the art recognized difficulties realized in the impact-modification polyester art is the unpredictability of the modifier effect, i.e., a modifier that enhances the properties of one polyester such as polybutylene terephthalate (PBT) or a PBT resin blend, may have little property modification in a different polyester or polyester blend. For example, certain polyacrylate modifiers such as Acryloid KM-330, while capable of significantly improving the impact properties of a PBT/PC resin blend, will have much less effect on a PET/PC blend.

About 20% of all plastic bottles produced in the United States are made from PET and the recycling of these bottles prove to be a major source of PET. These bottles are conventionally prepared from a polyethylene terephthalate resin with a base cup of high density polyethylene (HDPE) resin which comprises about one-third of the weight of the PET.

Separation plants (recyclers) often crush the bottles as received, i.e., together with the base cups, finely grind the material, and separate the components, paper (from labels), aluminum (from cups), PE (from base cups) and PET, by various methods including air separation, washing, and flotation. Since the presence of PE in the PET is deleterious to the overall properties of the PET resin, especially the impact strength of the resin, extensive separation procedures are used to obtain a PE-free PET stream for most recycle molding end-uses.

It would be highly desirable to improve the property spectrum, especially the impact strength of PET/PE resin blends and thus obviate the need for the additional separation now deemed necessary by recyclers and PET molders.

### SUMMARY OF THE INVENTION

A graft copolymer rubber resin material has been found that significantly enhances on a broad spectrum the physical properties of polyethylene terephthalate/polyethylene/polycarbonate resin blends.

In accordance with this invention rubbery spine polymer of a certain EPDM having grafted thereon styrene-acrylonitrile monomeric material has been found to unexpectedly increase the impact properties of PET/PE/PC blends.

The compositions of this invention are suitable for the molding of tough, high impact products and can be prepared from blending PET/PE material prepared by grinding PET bottles together with HDPE base cups; polycarbonate resin; and the graft copolymer rubber resin material described herein thus obviating the need for recyclers to remove the PE from the recycled bottles in order to achieve engineering grade PET resin.

### DETAILED DESCRIPTION OF THE INVENTION

The present invention is based on a blend of a) polyethylene terephthalate resin, b) polyethylene resin; c) polycarbonate resin; and d) graft copolymer composition comprising EPDM-type rubber grafted with styrene-acrylonitrile monomers. These blends have high impact properties even at low polycarbonate levels.

Preparations of polyethylene terephthalates are well known in the art and a basic description is described, for example, in U.S. Patent Nos. 2,465,319 and 2,471,023.

With regard to the term "polyethylene resin", it should be noted that in the industry, the term has come to include a variety of copolymers in which several significant percents of butylene, isobutylene or 1-hexene, are included. In the present invention, the most preferred, because of its use as a protective lower cup exteriorly on PET bottles is a high density polyethylene having density of from about 0.93 to 0.97 g/cc.

Polycarbonate resins can be made by known processes and they are available commercially from many sources, e.g., General Electric Co., Pittsfield, MA, U.S.A. under the trademark LEXAN. In general, any of the aromatic polycarbonates described in U.S. Patent No. 4,034,016; 4,257,937 and/or 4,280,949 can be used, especially those including units derived from bisphenol-A.

The graft copolymer material (d) constituting the fourth component of the invention is based on a graft copolymer of resin-forming monomeric material on a rubbery unsaturated spine polymer of ethylene-propylene-non-conjugated diene (EPDM). The preferred non-conjugated diene is ethylidene norbornene.

The grafted monomeric resin comprises the monomers styrene (which also includes substituted styrenes) and acrylonitrile. The styrene:acrylonitrile monomer ratios are in the range of from 90:10 to 40:60; preferably from 90:10 to 50:50; and most preferably from 80:20 to 60:40.

It will be understood that in practice the product of the graft copolymerization process is actually a mixture of true graft of resin on rubber along with a certain amount of separate, ungrafted resin (that is, the grafting efficiency is not 100 percent; see U.S. Pat. No. 4,202,948, Peascoe incorporated herein by reference.)

The graft copolymer of the present invention is an ethylene-propylene-non-conjugated diene terpolymer upon which is grafted a "graft resin".

The method by which the graft copolymer is formed, that is, the method by which the styrene-acrylonitrile graft resin is grafted onto the ethylene-propylene-non-conjugated diene terpolymer is set forth in detail in U.S. Patents 3,489,821; 3,489,822; and 3,642,950. All three of these patents are incorporated herein by reference. It is emphasized that although these patents teach grafting of not only styrene-acrylonitrile but other monomers as well, it is submitted that the method utilized therein applies to the formation of the graft copolymers of the present invention.

The graft copolymers of the present invention include a graft resin constituency, that is a styrene-acrylonitrile concentration, comprising between about 5 percent and 75 percent by weight, based on a total weight of the graft copolymer. More preferably, the graft resin comprises between about 30 percent and 70 percent by weight, based on the total weight of the graft copolymer. Most preferably, the graft resin comprises between about 40 percent and 60 percent by weight, based on the total weight of the graft copolymer.

The terpolymer of the present invention to which the graft resin is attached is characterized by a weight ratio of ethylene to propylene in the range of between about 50:50 and 75:25. Further, the ethylene-propylene-non-conjugated diene terpolymer, the so-called "rubber" portion of the graft copolymer, possesses an intrinsic viscosity, as measured in tetralin at 135° C, in the range of between about 1.5 and 2.6. The Mooney Viscosity (ML-4 at 257° F) of the rubber portion is in the range of between about 30 to 100. Typically, the ungrafted rubber is defined by an Iodine number in the range of between about 4 and 30.

Generally, the proportion of the four polymeric ingredients may vary, as indicated in the following table, wherein the amounts are expressed in parts by weight per 100 parts of blend:

	Parts by Weight		
	Broad	Preferred	More Preferred
(a) Polyester resin	20-90	20-80	40-60
(b) Polyethylene resin	1-40	1-30	1-20
(c) Polycarbonate resin	5-60	15-50	20-40
(d) Graft copolymer	5-50	15-40	20-30

The total amount of the polycarbonate resin and the graft copolymer should generally be greater than 40% by weight of the PET/PE/PC/graft copolymer blend.

Preferred compositions tend to include a major proportion of the polyester resin component (a) which combines good moldability and good properties at elevated temperature, along with sufficient polycarbonate resin (c) and graft copolymer (d) to provide high notched impact strength.

Although it is possible to use modifier (d) levels above about 30%, such an increase would tend to decrease hardness, tensile strength and stiffness.

There is no specific upper limit for the level of polycarbonate (c). Blends containing 40% (or more) of (c) are capable of providing satisfactory results.

To prepare the blends of the invention, the PET resin component (a), the polyethylene resin component (b), the polycarbonate resin component (c), and the graft copolymer composition (d) are mixed together at

elevated temperature in conventional plastics mixing equipment, such as a twin screw extruder-mixer. Since the mixing procedure is not critical so long as there is sufficient shear to disperse the EPDM graft copolymer in the blend, if desired, any two of the components may be mixed together first, followed by mixing in of the third and fourth components sequentially, at the same time or as a blend.

One desirable mixing procedure is a two-step compounding process involving first working the graft copolymer composition (d) without the other ingredients (a), (b) and (c) under shearing and fluxing conditions, for example in a twin screw extruder-mixer. This disperses the EPDM within the resin contained in the graft copolymer composition to form an "inverted" composition in which the EPDM rubber is a discontinuous phase. The second step involves fluxing the inverted graft component (d) with a PET/PE blend, e.g. a PET/HDPE bottle-cap regrind (a + b) and the polycarbonate component (c) under lower shear conditions, for example in a twin screw or single screw extruder. In commercial practice these two steps can be combined in one extrusion operation, using an extruder having a first and second feedport; a section of the extruder downstream of the first feedport can be used to shear (invert) the graft copolymer (d) and a section downstream of the second feedport can be used to mix the graft with the other ingredients (a), (b) and (c).

The inverted graft composition (d) may also be pelletized and tumble blended with PET (a), PE (b) or PET/PE (a + b) and polycarbonate (c) pellets to form a physical mixture which can be fed into an injection molding machine or an extruder. In this case the plasticizing screw of the injection or extrusion machine can perform the second stage of the mixing during fabrication.

As indicated, the invention is based on the discovery that four-component blends of polyethylene terephthalate (PET) and polyethylene (PE) with polycarbonate (PC) and ethylene-propylene-ethylidene norbornene (EPDM) grafted with styrene-acrylonitrile resin have unexpectedly high impact strength as well as a broad spectrum of other highly desirable physical and chemical properties. This invention has particular utility for molders using recycled PET which oftentimes contains PE from the residual in the recycling stream of bottle base-caps.

The following examples will serve to illustrate and delineate the practice of the invention in more detail.

#### Comparative Examples 1-6

The following examples illustrate the impact results realized via the use of the polyacrylate impact modifier Acryloid KM-330 (a trademark of Rohm and Haas) in polyethylene terephthalate/polycarbonate and polybutylene terephthalate/polycarbonate blends.

The blends listed in Table 1 are prepared by fluxing the Acryloid KM-330 with PET (KODAPAK 7352; trademark EASTMAN CHEMICALS) or PBT (VALOX 310; trademark of General Electric Co.); and polycarbonate resin (reaction product of bis-phenol A with phosgene (LEXAN 141; a trademark of General Electric Co.) in a 1" single-screw extruder equipped with strand die and pelletizer. Specimens for mechanical property testing are cut from 1/8 inch injection molded plaques.

In Table 1 (and the remaining Tables) the amounts of each component are expressed in parts by weight; PET stands for the polyethylene terephthalate; PBT stands for the polybutylene terephthalate; PC stands for the polycarbonate resin; NIRT stands for Notched Izod Impact (ASTM D256) strength at room temperature (reported in foot pounds per inch of notch). NIRT (annealed) means Notched Izod Impact strength at room temperature determined after annealing the specimen for 2 hours at 150° C in a circulating air oven. The annealing process causes the PET to crystallize and will tend to increase tensile and stiffness of the blend. The PBT blends are crystallized as molded.

TABLE I

Comparative Examples	1	2	3	4	5	6
Acryloid KM-330	10	20	30	10	20	30
PET				70	60	50
PBT	70	60	50			
PC	20	20	20	20	20	20
NIRT (As molded)	3.1	21.2	17.8	1.5	14.9	11.7
NIRT (Annealed)	--	--	--	1.0	2.0	2.2

The above comparative examples show that although a given polyacrylate additive may perform satisfactorily as an impact modifier for certain polyester/polycarbonate blends, specifically PBT/PC blends, one cannot predict that the additive will affect the impact properties of another polyester, albeit a homologous one, in a similar manner.

Comparative Examples 7-14

Three graft copolymers, designated A, B and C in TABLE II below, are prepared by the slurry technique described in U.S. Patent No. 4,202,948 (Peascoe) using an ethylene-propylene-ethylidene norbornene EPDM terpolymer rubber (EP ratio 50/40; Mooney viscosity 58 ML-4 @ 257° F; iodine number 20) as the spine rubber. The resin forming monomers in each graft are indicated in TABLE II, which gives the composition of the resin portion of the graft copolymers. The overall resin/EPDM ratio in the graft copolymer is 50/50 in each case.

TABLE II

	Resin	Ratio
A	Styrene/Acrylonitrile	72/28
B	Styrene/Methyl Methacrylate	50/50
C	Methyl Methacrylate	100

The blends listed in TABLE III are prepared by first fluxing the graft copolymers (A, B or C) in a 53 mm Werner and Pfleiderer twin-screw extruder equipped with a strand die and pelletizer. The pelletized grafts are then fluxed with PBT (VALOX 310; trademark of General Electric Co.) and polycarbonate resin (reaction product of bisphenol A with phosgene; LEXAN 103 (trademark of General Electric)) in a one-inch single-screw extruder to make the blend shown in TABLE III. Specimens for mechanical property testing are cut from 1/8 inch injection molded plaques. (See U.S. Patent No. 4,493,921).

TABLE III

Comparative Examples	7	8	9	10	11	12	13	14
B	20			10	30		20	
C		20						
A			20			30		20
PBT	60	60	60	70	50	50	40	40
PC	20	20	20	20	20	20	40	40
NIRT	17.8	15.0	17.9	2.6	19.2	19.0	16.4	16.2

Exmaples 15-24

5 Th following examples in Table IV show the deleterious impact property results that would be realized in molded articles by the inclusion of PE resin, either as an added component or as a residual in PET/PC resin blends and the outstanding impact property improvements achieved via the incorporation of the rubber graft copolymers of this invention.

10 The term "polyethylene terephthalate regrind" refers to PET resin obtained from the recycling of PET articles, primarily bottles. The term "polyethylene regrind" refers to PE resin obtained from the recycling of PE articles, primarily bottle base-cups. The "PET Regrind" is obtained from NYQONN Industries and is a PET soda bottle recycle product. The "PE Regrind" is obtained from M.A. Industries and is a PE soda bottle base-cup recycle product.

The term "polyethylene terephthalate/polyethylene regrind" refers to PET/PE resin blends obtained from the recycling of PET/PE articles, primarily PET bottles and the PE base-cups affixed thereon.

15 The polycarbonate (PC) resin (reaction product of bisphenol A with phosgene) is LEXAN 141 (trademark of GE). The Gardner Drop Weight test (DWI) utilizes a six (6) pound weight; 5/8" hemispherical striker; and a 1 1/2" support on a 1/8" thick plaque. Tensile properties are measured according to ASTM D638.

The EPDM graft F is prepared by the slurry technique described in U.S. Patent No. 4,202,948 (Peascoe) and uses an ethylene-propylene-ethylidene-norbornene (EPDM) terpolymer rubber (E/P ratio 20 60:40; Mooney viscosity 68 ML-4 at 257° F, iodine number 20) as the spine rubber. The resin of graft copolymer F is a styrene-acrylonitrile resin with a ratio of 72:28. The weight ratio of EPDM to resin is 50:50.

25 The blends listed in Table IV are prepared by first fluxing the graft copolymer in a 53 mm Werner and Pfleiderer twin-screw extruder equipped with a strand die and pelletizer. The pelletized grafts are then fluxed with the other ingredients in a 30 mm ZSK twin screw extruder to make the blends. Specimens for mechanical property testing are prepared by injection molding.

TABLE IV

Examples	15	16	17	18	19	20	21	22	23	24
EPDM Graft F				30	30	30	25	17.5	30	
Acryloid KM-330				37.5	37.5	45	50	50	50	30
PET Regrind	100	50	50	12.5	12.5	5	5	12.5	50	50
PE Regrind			30	20	20	20	20	20	20	20
Polycarbonate		50	20	20	20	20	20	20	20	20
NIRT (t-ibfn) (as molded)	1.0	2.0	1.5	8.1	15.8	19.2	17.4	5.1	22.5	18.4
NIRT (t-ibfn) (annealed)	0.8	1.3	1.1	4.1	5.6	6.1	3.6	1.2	22.0	4.5
DWI (n-ib)* (as molded)	75-240NB	240NB	<3	12	240NB	240NB	240NB	75-240NB	240NB	240NB
DWI (n-ib)* (annealed)	12	240NB	<3	12	240NB	240NB	240NB	30-100	240NB	240NB
Tensile (psi) (as molded)	8853	8942	5281	4278	4788	5321	5704	5780	5345	5020
Tensile (psi) (annealed)	12,007	10,546	5928	4908	5773	6253	8783	7127	6202	8134

\* Lower value near gate end.

\*\* NB = No Break (i.e., exceeds capacity of machine)



Examples 19 through 22 in Table IV illustrate the outstanding improvements realizable in the impact properties of PET resin compositions which contain PE resin such as is the case with the PET/PE blends produced when PET soda bottles are recycled together with the PE base cups. Examples 19 is of special note for the PET/PE ratio approximates that found in PET/PE soda bottle recycle grind.

Additionally, comparison of the results realized in Examples 19, 20 and 21 versus Example 22 illustrates the more greatly enhanced properties achieved when the weight percent of the polycarbonate and the graft copolymer exceeds 40% of the total four component blend.

Particular note should be taken of the significant drop in impact properties (as measured by DWI) that occurs with the addition of PE to the PET/PC blends, i.e., Example 17 versus Example 16, in the absence of the graft modifiers of this invention. Furthermore, although the Acryloid KM-330 modifier is effective in realizing good impact properties in PE/PC blends (Ex. 24) it is relatively ineffective as an impact modifier when PE is present (Ex. 18).

#### Examples 25-34

Two graft copolymers, designated D and E are prepared by the slurry technique described in U.S. Patent No. 4,202,948 (Peascoe). Graft copolymer D used an ethylene-propylene-ethylidene-norbornene EPDM terpolymer rubber (EP ratio 60/40; Mooney viscosity 68 ML-4 at 257° F, iodine number 20) as the spine rubber. Graft copolymer E used an ethylene-propylene-dicyclopentadiene EPDM terpolymer rubber (EP ratio 52/48; Mooney viscosity 60 ML-4 at 257° F, iodine number 7) as the spine rubber. The resin forming monomer in the grafts of D and E is methyl methacrylate with a ratio of 72/28. The weight ratio of EPDM to resin is 50/50.

The blends listed in TABLE V are prepared by first fluxing the graft copolymers (D, E or F) in a 53 mm Werner and Pfleiderer twin-screw extruder equipped with a strand die and pelletizer. The pelletized grafts are then fluxed with PET (KODAPAK 7352; a trademark of Eastman Chemicals) and polycarbonate resin (reaction product of bis-phenol A with phosgene (LEXAN 141; a trademark of General Electric Co.)) in a one-inch single screw extruder to make the blends. Specimens for mechanical property testing are cut from 1:8 inch injection molded plaques.

TABLE V

EXAMPLE	25	26	27	28	29	30	31	32	33	34
D	20				20			20		
E		20				20			20	
F			20	30			20			20
PET	60	60	60	50	50	50	50	40	40	40
PC	20	20	20	20	30	30	30	40	40	40
NIRT (as molded)	3.0	1.9	10.2	18.7	18.4	15.6	19.3	19.5	16.0	19.1
NIRT (annealed)	0.8	0.7	0.8	15.2	4.6	5.3	4.8	15.8	12.1	17.7

TABLE V illustrates the overall superiority of Notched Izod Impact for graft F, containing styrene-acrylonitrile as the grafted resin (Examples 27, 28, 31 and 34).

#### Examples 35-44

Two additional graft copolymers, designated g and H were prepared similarly to graft copolymers D and F except that the resin forming monomer in G was methyl methacrylate with an EPDM/resin ratio of 47/53 and the resin forming comonomers in H was styrene-methyl methacrylate (50/50) with an EPDM/resin ratio of 47/53.

The blends of TABLE VI were prepared as those in Examples 15-24 and specimens were similarly prepared.

TABLE VI

EXAMPLE	35	36	37	38	39	40	41	42	43	44
G	30					20				
H		30					20			
D			30					20		
E				30					20	
F					30					20
PET	50	50	50	50	50	40	40	40	40	40
PC	20	20	20	20	20	40	40	40	40	40
NIRT (annealed)	8.6	6.8	14.3	11.9	15.2	12.7	16.2	15.8	12.1	17.7

The above TABLE VI underscores the superior results generally realized via the graft resins of this invention in PET/PC blends (Examples 39 and 44).

The foregoing examples of this invention may be repeated using other aromatic polycarbonate resins; polyethylene terephthalate resins; polyethylene resins or ethylene-propylene-non-conjugated diene terpolymers grafted as set forth above.

#### Claims

##### a. A blend comprising

- polyethylene terephthalate resin;
  - polyethylene resin;
  - polycarbonate resin; and
  - graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-non-conjugated diene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 5-75% by weight of the graft copolymer; and
- wherein the proportions of (a), (b), (c) and (d) expressed in parts by weight per 100 parts of (a), (b), (c) and (d) are as follows:
- 20-90;
  - 1-40;
  - 5-60; and
  - 5-50.

##### 2. A blend comprising

- polyethylene terephthalate resin;
  - polyethylene resin;
  - polycarbonate resin; and
  - graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-ethylidene norbornene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 5-75% by weight of the graft copolymer; and
- wherein the proportions of (a), (b), (c) and (d) expressed in parts by weight per 100 parts of (a), (b), (c) and (d) are as follows:
- 20-90;
  - 1-40;
  - 5-60;
  - 5-50.

##### 3. A blend comprising

- polyethylene terephthalate resin;
  - polyethylene resin;
  - polycarbonate resin; and
  - graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-ethylidene norbornene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 5-75% by weight of the graft copolymer, said graft copolymer being an inverted composition in which the rubber is a discontinuous phase; and
- wherein the proportions of (a), (b), (c) and (d) expressed in parts by weight per 100 parts of (a), (b), (c) and (d) are as follows:

- (a) 20-80;  
 (b) 1-30;  
 (c) 15-50; and  
 (d) 15-40.
- 5 4. A blend comprising
- a) polyethylene terephthalate resin;  
 b) polyethylene resin;  
 c) polycarbonate resin; and  
 d) graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-ethylidene norbornene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 30-70% by weight of the graft copolymer, the graft copolymer having been worked under shearing and fluxing conditions to disperse the rubber portion of the graft within the resinous portion thereof thus forming an inverted composition in which the rubber portion is a discontinuous phase prior to blending with (a), (b) and (c); and wherein the proportions of (a), (b), (c) and (d) expressed in parts by weight per 100 parts of (a), (b), (c) and
- 15 (d) are as follows:  
 (a) 20-90;  
 (b) 1-40;  
 (c) 5-60 and  
 (d) 5-50.
- 20 5. The blend of claim 1 wherein the proportions of (a), (b), (c) and (d) are as follows:  
 (a) 20-80;  
 (b) 1-30;  
 (c) 15-50 and  
 (d) 15-40.
- 25 6. The blend of claim 1 wherein the proportions of (a), (b), (c) and (d) are as follows:  
 (a) 40-60;  
 (b) 1-20;  
 (c) 20-40, and  
 (d) 20-30.
- 30 7. A method of making a blend of
- a) polyethylene terephthalate resin;  
 b) polyethylene resin;  
 c) polycarbonate resin; and  
 d) graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-ethylidene norbornene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 30-70% by weight of the graft copolymer, and wherein the proportions of (a), (b), (c) and (d) expressed in parts by weight per 100 parts of (a), (b), (c) and
- 40 (d) are as follows:  
 (a) 20-90;  
 (b) 1-40;  
 (c) 5-60; and  
 (d) 5-50
- 45 comprising:  
 working the graft copolymer (d) under shearing and fluxing conditions to disperse the rubber portion of the graft within the resinous portion thereof, thus forming an inverted composition in which the rubber portion is a discontinuous phase, and thereafter blending the resulting inverted graft with components (a), (b) and (c).
8. A method of making a blend according to claim 7 wherein said styrene-acrylonitrile resin is present from about: 30-70% by weight of the graft copolymer and the proportions of (a), (b), (c) and (d) are as follows:  
 (a) 20-80;  
 (b) 1-30;  
 (c) 15-50; and  
 (d) 15-40.
- 55

9. A blend comprising

- a) polyethylene terephthalate regrind;
  - b) polyethylene regrind;
  - c) polycarbonate resin; and
  - 5 d) graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-non-conjugated diene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 5-75% by weight of the graft copolymer, and
- wherein the proportions of (a), (b), (c) and (d) expressed in parts by weight per 100 parts of (a), (b), (c) and (d) are as follows:
- 10 (a) 20-90;
  - (b) 1-40;
  - (c) 5-60; and
  - (d) 5-50.

10. A blend comprising

- 15 a) polyethylene terephthalate; polyethylene regrind;
  - b) polycarbonate resin; and
  - c) graft copolymer of styrene-acrylonitrile resin on an ethylene-propylene-non-conjugated diene rubber spine, said styrene-acrylonitrile resin being present in an amount of about 5-75% by weight of the graft copolymer, and
- 20 wherein the proportions of (a), (b), and (c) expressed in parts by weight per 100 parts of (a), (b) and (c) are as follows:
- (a) 20-90;
  - (b) 5-60; and
  - 25 (c) 5-50.

12

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71 Applicant: **UNIROYAL CHEMICAL COMPANY,  
Inc.**  
**World Headquarters**  
**Middlebury Connecticut 06749(US)**

72 Inventor: **Wefer, John Michael**  
**Saddle Ridge Road Newtown**  
**Fairfield Connecticut 06470(US)**

74 Representative: **Browne, Robin Forsythe et al**  
**Urquhart-Dykes & Lord Tower House Merriam**  
**Way**  
**Leeds LS2 8PA West Yorkshire(GB)**

54 **Impact resistant polyethylene terephthalate/polycarbonate/PE blends.**

57 High impact strength blends based on polyethylene terephthalate resin, polyethylene resin, aromatic polycarbonate resin and styrene-acrylonitrile grafted copolymers of EPDM. This invention is of special value in improving the engineering properties of PET/PE bottle recycle products.

**EP 0 322 095 A3**



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# EUROPEAN SEARCH REPORT

Application Number

EP 88 31 0229

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. Cl.4)
Y	EP-A-0 135 677 (GENERAL ELECTRIC CO.) * Claims 1-2,4-8; page 5, lines 30-35 *	1-2	C 08 L 67/02 C 08 L 23/06 C 08 L 69/00 C 08 L 51/06 C 08 J 11/06
Y	EP-A-0 110 222 (GENERAL ELECTRIC CO.) * Claims 1,3,9-12,14-17,21; page 13, lines 13-26 *	1-2	
P,A	EP-A-0 287 207 (UNIROYAL CHEMICAL CO.) * Claims 1-10 *	1-8	
A	EP-A-0 115 015 (GENERAL ELECTRIC CO.) * Claims 1-2,7,12-19,23; * & US-A-4 493 921 (Cat. D)	1-3	
A	EP-A-0 107 303 (UNIROYAL, INC.) * Claims 1,3-8; page 2, lines 15-35: page 3, lines 5-34 *	1-8	
			TECHNICAL FIELDS SEARCHED (Int. Cl.4)
			C 08 J C 08 L
The present search report has been drawn up for all claims			
Place of search THE HAGUE		Date of completion of the search 10-10-1990	Examiner MIAO K. Y-P.
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons & : member of the same patent family, corresponding document	